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DAMAGE RESISTANCE OF COATED OPTICS FOR PULSED, CO_2 LASERS*

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Abstract

The influence of various factors on the damage threshold of coated optical components used in high-power, pulsed CO_2 lasers is reviewed. The factors considered are substrate roughness and polishing ~~defects~~, coating defects, linear absorption, deposition parameters, standing-wave electric fields, pulsewidth dependence, multiple-shot conditioning, and optical performance. Experimental results of many researchers are used to illustrate the discussions.

Introduction

The damage thresholds of coated optical components are frequently the major design and operating constraints on pulsed, high-power CO_2 lasers. There are a variety of CO_2 laser components which employ thin-film coatings and these include: total reflectors, partial reflectors, gratings, windows, saturable absorbers, Faraday isolator rods, modulator and Q-switch crystals, polarizers, waveplates, and lens. Of these, all but the first three require anti-reflection (AR) coatings. Thus, in the following discussions, aspects of laser damage which impact AR coatings in particular will be emphasized. The topics to be considered generally apply to coated optics for all laser wavelengths, but the experimental data for CO_2 lasers will be used to illustrate our present knowledge of this subject. Furthermore, a slight emphasis will be placed on data for pulsewidths of 100 ns or less for which peak intensity considerations are important. The following topics will be reviewed: 1) substrate roughness and polishing defects, 2) coating defects, 3) linear absorption, 4) deposition parameters, 5) standing-wave (SW) electric fields, 6) pulsewidth dependence, 7) multiple-shot conditioning, and 8) optical performance.

Substrate Roughness and Polishing Defects

A primary cause of early failure of coated interfaces through which intense pulses are transmitted is the substrate finish. It is commonly known that the damage threshold of the bare surface of a laser component is generally much lower than that of the bulk interior. N. Bloembergen[1] has correctly identified the reason for this difference to be due to the polishing process which produces a microscopically rough surface with randomly distributed cracks and grooves, and which leaves embedded residual amounts of polishing compound. Representative geometries of such defects are illustrated in Figure 1. The interaction of the incident laser electric field with the imperfect surface results in enhancement of the field at these defect sites. The magnitudes of the enhancement at the sites A, B, and C are given by the equations in Table 1; values for CO_2 laser substrate materials are listed in Table 2.

Table 1. Electric-Field Enhancement[1]

$$E_{ins} = \frac{1}{1 + \frac{1-\epsilon}{\epsilon}L} E_o$$

L = depolarization factor

A. Sphere	B. Cylinder	C. Crack
$L = 1/3$	$L = 1/2$	$L = 1 - \frac{\pi}{2}\frac{c}{a}$
		~ 1 for
$E_{ins} = \frac{3\epsilon}{2\epsilon+1} E_o$	$E_{ins} = \frac{2\epsilon}{\epsilon+1} E_o$	$c/a \ll 1/\epsilon < 1$
		$E_{ins} = \epsilon E_o$

*Work performed under the auspices of US DoE

Table 2. Electric-Field Enhancement Factors for CO_2 Laser Substrates

Material	Sphere	Cylinder	Crack
NaCℓ n=1.5	1.23	1.38	2.25
ZnSe n=2.4	1.38	1.70	5.76
CdTe n=2.69	1.40	1.76	7.24
Ge n=4.0	1.45	1.88	16

It is obvious that incipient-crack defects cause the greatest enhancement, dramatically so for high-index materials. Since it is the interaction of the electric field which causes the damage to the surface via absorption or electron avalanche, the defect sites are susceptible to early damage. In terms of the incident laser intensity, the damage threshold is reduced by the square of the enhancement factor. Thus, for a crack in a Ge surface, the threshold would be degraded by a factor of 256.

The implications for coated surfaces are just as serious. From microscopic examination, it is known that most coatings faithfully replicate the contour of the substrate. So, the coating will experience enhanced electric fields also. Either the coating will itself damage at a lower intensity or the substrate will be damaged with resultant coating delamination. Experimental evidence relating the breakdown field of a coated surface to the substrate roughness has been obtained by House and coworkers.[2] In Figure 2 a plot of the field versus substrate roughness for half-wave SiO_2 films on fused silica substrates produced the relation $E \sim \sigma^{-m}$, where m is 0.42. Although the experiments were conducted with a 1.06 μm laser on SiO_2, the same relationship is applicable to other wavelengths and materials with appropriate values of the exponent m.

Recently, the effect of three different polishing techniques on the damage threshold of AR-coated Ge was measured at 10.6 μm at Los Alamos Scientific Laboratory. The AR coatings were deposited in the same run. The results were that two of the windows with visibly obvious polishing scratches failed at 70% of the threshold of the third window which exhibited no visible polishing marks.

There are at least two ways to decrease the roughness of substrates used for infrared windows. One is to etch away the rough surface layer by use of an acid treatment or by use of ion beams. The latter treatment has been found to raise the damage resistance of Ge surfaces significantly.[3] A second method is to deposit an initial or "barrier" layer which does not faithfully replicate the substrate profile but presents a smoother surface for the next coating layer. Such an effect was produced by depositing a layer of CeF_3 on a ZnS substrate.[4] Also, an amorphous coating of As_2S_3 is surmised to do likewise. A half-wave layer of this material deposited on a KCl window dramatically increased the damage threshold by a factor of two.[5]

Coating Defects

Early failure of coatings with low absorption coefficients has been attributed to coating defects. These defects can take the form of absorbing inclusions, voids, and pinholes ranging from submicron diameters to visually discernible dimensions. Gibbs and Wood[5] have measured the damage thresholds of areas free from visible defects and those containing defects on the same sample. Their results, listed in Table 3, indicate that visible defects can reduce the damage threshold by a factor of two to three for AR coatings and partial reflectors and by a much larger factor for total reflectors.

One of the indicators of the presence of coating defects is the experimentally measured dependence of the threshold on the area of irradiation. It is reasonable to expect that large diameter beams, on the average, will damage materials more easily than small diameter beams. This follows from 1) the greater probability of striking a randomly distributed defect when a larger area is irradiated and 2) the fact that materials with defects damage more easily than those without defects. Figure 3 presents the spot-size dependence of multilayer dielectric (ThF_4/ZnS)-enhanced Mo relfectors.[6] With due caution, such a plot can be used to obtain a rough estimate of the number of defects per unit area which are susceptible to damage.

Table 3. Effect of Coating Defects on Damage Threshold[7]

Description	Damage Threshold (MW/cm^2) Defect	Clear Area
Single-layer AR		
ZnS on Si	40	120
ZnS on Ge	140	280
ZnS on Ge	280	400
ThF_4 on ZnSe	300	>600
R=85%		
ZnS/ThF_4 on Ge	500	1000
R≈100%		
ThF_4/ZnS on SiO_2	80	380
on Si	<20	120

Laser irradiation: TEA CO_2, 60 ns (FWHM) initial pulse plus 700 ns tail, 0.070 mm^2 spot-size radius.

For vanishing spot-size the threshold represents the intrinsic limit of a defect-free coating. For larger spot-sizes, the defect-limited threshold is measured and this, of course, is the practical value which applies for use in a laser system. When comparing damage thresholds measured by different investigators, it is important to verify that the spot-size radii are either the same or conservatively large, e.g., greater than 0.25 mm.

Linear Absorption

It is appropriate for the coating manufacturer to minimize the magnitude of linear absorption in a coating. Absorption decreases the main function of the coating, to transmit or reflect, and causes thermal loading. If a large enough temperature excursion results, wavefront distortion or film delamination or rupture can result. However, for laser pulses shorter than 100 to 200 ns, it appears that linear absorption of a coating is not of primary importance in determining its damage resistance.

In Table 4, the damage thresholds of several QW low-index and HW high-index coatings can be compared with their absorption coefficients. For the 60-ns pulse irradiation, the thresholds are not inversely proportional to absorption as might be expected, but the opposite is apparent. Further, as shown in Table 5, the thresholds for 1.15 ns laser pulses incident on identical AR coatings of ThF_4/ZnS on GE (four different substrates with identical surface polish) are not strongly correlated with substrate absorption. A likely explanation is that either heating of tiny coating defects or electron avalanche is the limiting process for short laser pulses. The high electric fields present in short pulses can produce the approximately $10^{18}/cm^3$ free electrons required for the avalanche process prior to significant linear absorption within the film or substrate.

Table 4. Short-Pulse Damage Threshold Versus Film Absorption at 10.6 μm[7]

Coating/window	Coating Index	Absorption Coefficient (cm^{-1})	Energy Density (J/cm^2)
SrF_2/KCl	1.36	25	194
$BaF_2/NaCl$	1.38	15	76
ThF_4/KCl	1.35	10-12	
$NaF/NaCl$	1.23	6.0	38
$As_2S_3/NaCl$	2.37	1.2	38
$As_2Se_3/NaCl$	2.72	1.1	12

Laser irradiation: Single-mode CO_2, 60 ns (FWHM) pulse, 0.10 mm spot-size radius.

Table 5. Short-Pulse Damage Threshold of AR-Coated Ge Versus Substrate Absorption[8]

Coating: s/ThF_4/ZnS/air

Germanium Substrate	Absorption Index (cm^{-1})	Energy Density J/cm^2
Single Crystal (undoped)	0.005	0.47
Single Crystal(Ga-doped)	0.6	0.47
Polycrystal (undoped)	0.005	0.48
Polycrystal (Ga-doped)	0.6	0.41

Laser irradiation: CO_2-P(20), 1.15 ns pulse (FWHM), 1.1 [illegible] spot-size radius.

For long-pulse irradiation, coating damage does appear to be directly proportional to absorption. This is confirmed by the data in Table 6 for a variety of coatings subjected to a 200+ ns CO_2 laser pulse. Apparently, damage by film heating preceeded the formation of an electron avalanche.

Table 6. Long-Pulse Damage Threshold versus Film Absorption at 10.6 μm

Coating	Absorption of coating	Damage Threshold J/cm^2
	a. QW Films on KCℓ[9]	
ThF_4	10 cm^{-1}	80
As_2S_3	< 1 cm^{-1}	120
ZnSe	< 0.6 cm^{-1}	140
	b. QW ThF_4 on Cu[10]	
ThF_4	8.6% total	1.0
ThF_4	2.7%	2.5
	c. AR coats on KCℓ[5]	
As_2S_3/ThF_4	0.80%	6-10
As_2S_3/ThF_4	0.19%	31

Laser irradiation: TEA CO_2, 200 ns (FWHM) primary pulse with 3 μs tail with multilongitudinal mode beating; spot-size radius of a) 35 μm, b) 1mm and c) 200 μm

In summary, it is believed that in the absence of homogeneous film absorption or localized absorption by defects in a coating the electron avalanche process would be the damage mechanism. When significant linear absorption occurs, however, deposition of sufficient energy density over long pulse periods to cause coating damage will occur before the onset of an avalanche.

Coating Deposition Parameters

As coating manufacturers know, the combination of deposition parameters must be optimized to obtain films with minimum linear absorption. These parameters include substrate cleaning, starting-material chemical state and purity, evaporation method, substrate temperature, residual gases in the vacuum chamber, and evaporation rate. As an example, a few years ago researchers at Hughes Research Laboratories determined the optimum coating parameters for the widely-used low-index material, ThF_4. The typical attainable absorption coefficients had ranged from 10 to 20 cm^{-1}, but Wang and coworkers reported finding conditions that obtained a value near 1 cm^{-1}.[6] This was accomplished by minimizing the residual water in the vacuum chamber. Long pumpdowns at 10^{-7} Torr and a substrate temperature of 150°C were required. Additionally, the starting material was a crystal chunk free of major impurities. This material was evaporated by a resistance-heated tantalum box source at a deposition rate of 630 nm/min.

Standing-wave Electric Fields

Standing-wave electric fields are the basis of the operation of thin film coatings, and the distribution and magnitude of the maxima and minima within a coating bear is related to the normalized electric field by

$$P_a = n\alpha \left| E/E_o^+ \right|^2 P_o \qquad (1)$$

where α is the absorption coefficient and P_o is the incident power density. In the case of electron avalanche, the growth of the number density with time is given by

$$N(t) = N_o \int_o^t \exp\left[\alpha(E)\right] dt, \qquad (2)$$

where α is the gain coefficient. Since both damage mechanisms are functions of the eletric field, coating designs with reduced field maxima in weak or absorbing layers are advantageous. Knowledge of the field distribution also enables one to predict the relative threshold of a coated surface.

An example is the recent experimental evaluation of AR coatings on Ge by Newnam and Gill.[8] Their results, summarized in Table 7, indicated that the damage resistance of 14 coating designs using eight different film materials were effectively the same, 0.49 + 0.03 J/cm³, and were lower than uncoated Ge. Furthermore, damage occurred only at the front surface. The results were explained by considering the electric-fields in the Ge, coated and uncoated as shown in Figure 4. Although the exact field distributions within the various coating designs differed quantitatively, the gradual decrease from 1.0 at the air-film interface to 0.25 at the film-Ge interface was the same for all designs. The ratio of the fields-squared in uncoated-to-coated Ge was therefore 0.64. Since the ratio of the measured thresholds was 0.7 + 0.1, which is consistent with the field ratio, it was concluded that the damage occurred in the Ge at the Ge/film interface.

Table 7. Damage Thresholds of AR-Coated and Uncoated, P-Doped, Polycrystalline Ge[8]

(Front Surface only)

	Energy Density (J/cm^2)
AR-coated Ge	0.49 ± 0.03 (average of 14 designs)
Bare Ge p-doped	0.64 ± 0.05

Laser irradiation: CO_2 P(20), 1.12 ns pulse (FWHM), 1.1 mm spot-size radius.

Consideration of the SW field distributions in various AR coating designs for substrates with wide bandgaps, e.g. KCl, suggests a three-layer design. In this design, the low-index film is deposited as the middle layer where the field is at a minimum. This minimizes the linear absorption in this layer which, for long pulsewidths, is more susceptible to thermally-induced damage than the high-index components. Baer, et al.[7], produced such a coating of $As_2S_3/ThF_4/As_2S_3$ which had half the absorption of a two-layer AR coating using these same materials.

Pulsewidth Dependence

The damage threshold of optical materials, in terms of Joules/cm² increases with increasing laser pulsewidth for several reasons. With increasing pulsewidths, nonlinear absorption is reduced due to lower power densities, and heat and electrons have more time to diffuse away from the irradiated surface. The experimentally confirmed relationship for the damage threshold of metals and metal films of sufficient thickness is

$$\mathcal{E} = Kt^{\frac{1}{2}}, \ (J/cm^2), \qquad (3)$$

where K is a constant comprising the thermal properties and absorption of the metal. For Cu, for example, the theoretical value of K is 4.4×10^5 J/cm² - sec$^{\frac{1}{2}}$ for a wavelength of 10.6 μm.[11] This $t^{\frac{1}{2}}$ power dependence has also been measured for AR-coated and uncoated GE surfaces at the CO_2 wavelength for pulsewidths from 1 to 70 ns as seen in Figure 5. This similar dependence to that of a metal is believed to result from the laser-induced generation of free electrons at easily ionized defect sites at the Ge surface. The $t^{\frac{1}{2}}$ power dependence has also been measured for optical breakdown in the bulk of NaCl for pulsewidths from 15 ps to 10 ns as shown in Figure 6.[12,13,14] Although the measurements were conducted with a 1.06 um laser, the observed pulsedwidth dependence is also applicable to the CO_2 laser wavelength, since the damage thresholds of alkali halide crystals for a given pulsewidth have been measured to be equal from 0.7 to 10.6 um.[15]

The $t^{1/2}$ dependence of the energy-density threshold is due to the existing (in metals) and laser-generated (in dielectrics) free-electron plasma which absorbs the remaining pulse energy which is transferred to and damages the immediate solid. For absorption of pulsed energy at absorbing defect sites or in homogeneously absorbing coatings, it is expected that the damage threshold would increase more slowly than $t^{1/2}$ due mainly to thermal diffusion. In particular, the thresholds for a multilayer-enhanced Mo reflector measured by Wang, et al.,[6] at long pulselengths show no dependence on pulsewidth (see Table 8).

For even longer pulsewidths, e.g. 100 µs, it is expected that the damage threshold would increase slowly depending on the rate of thermal diffusion.

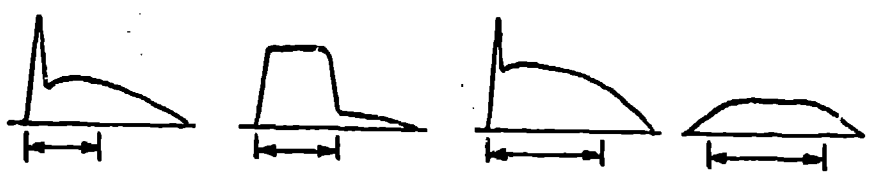
Table 8. Single - and Multiple-shot Damage Threshold of an Enhanced

Mo Reflector for Four Long CO_2 Pulseforms [6]

Reflector: $Mo/A_g/(ThF_4/ZnSe)^4$
Absorption: 0.30%
Spot-size radius = 70 µm

Damage Threshold (J/cm^2)

	0.6 µs Mode locked	1 µs Single-long.mode	4 µs Mode-locked	.6 µs Single-long.mode
Single-shot Threshold	15-75	~70	70-110	50-90
Multiple-shot Threshold	50-290	110-460	110-200	70-290



0.6 µs   1 µs   4 µs   6 µs

Multiple-Shot Conditioning

Several investigators have reported that sites in a sample which have been pre-irradiated by low laser intensities have higher damage thresholds than unconditioned sites. At surfaces and coatings, it has been speculated that some sort of cleaning or outgassing of contaminants occurs in the conditioning process. Such an increase, by a factor of two or more, is seen in the multiple-shot thresholds presented in Table 8. At a shorter pulsewidth of 1.2 ns, the author has observed a similar but smaller (20 to 50%) increase in the damage threshold of AR-coated and uncoated NaCl. However, no conditioning effect, positive or negative, was observed in multiple-shot tests on AR-coated Ge.[8] In this case, the damage-prone surface was beneath the coating at the substrate interface. Additional research is needed to clarify this issue.

Optical Performance

One question often asked by laser users is how far below the single-shot damage threshold a component can be irradiated and survive for a specified number of shots without significant degradation of its performance. Gill and Newnam[16] have performed such performance experiments on AR-coated ZnSe and NaCl windows using 1.2 ns CO_2 laser pulses. Since focusable energy on a target is of primary importance to laser fusion systems, the relative amount of energy focusable through a small aperture after transmission through the test window was the quantity measured. Figures 7 and 8 are plots of the focusability as a function of shot number for several irradiation levels above and below the single-shot thresholds (SST) 1.2 and 4.3 J/cm^2, respectively, for the two windows. For both windows, irradiation levels of 50 to 60% SST produced no degradation after 90 shots. At greater irradiation levels, the focusability was seen to decline. The gradual decline for the ZnSe window in contrast to the abrupt fall off for the NaCℓ window is due to its finer-grained polycrystalline structure. Finally, the effective lifetime for 80% focusability as a function of irradiation fluence is presented in Figure 9. Such information is very important for the laser system designer.

Summary

We have reviewed eight factors which influence the damage resistance of coated optics for pulsed CO_2 lasers. Other aspects such as film stress and coating-to-substrate adhesion have not been considered here and need similar attention in a thorough review. From the evidence presented, it is especially interesting how the relative importance of linear absorption and avalanche breakdown changes with pulselength. The former is dominant for pulsewidths longer than ~200 ns, the latter for shorter times. In each of these time regimes, coating and substrate defects limit the attainable thresholds considerably below the intrinsic limits. With systematic, concerted effort, practical methods to decrease the defect limitations are certain to be developed.

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Figures

Fig. 1. Representative geometries for electric-field enhancement near pores, scratches and incipient cracks. Typical dimensions are r = 0.1 μm, c = 0.1 μm and a = 1 μm (after Bloembergen).[1]

Fig. 2. Electric field E (~$\sqrt{\varepsilon}$) threshold versus roughness of the fused silica substrate for half-wave SiO_2-overcoated samples. Laser irradiation: 1.06 um, 40 ns pulse (FWHM), 0.15 mm spot-size radius (after House, et al.).[2]

Fig. 3. Single-shot damage thresholds as a function of spot size for seven 6- and 8-layer ThF_4/ZnSe enhanced Mo reflectors. Laser irradiation: TEA CO_2, smooth 6 us, single-longitudinal-mode pulse (after Wang, et al.).[6]

Fig. 4. Standing-wave electric fields for AR-coated and uncoated Ge substrates.

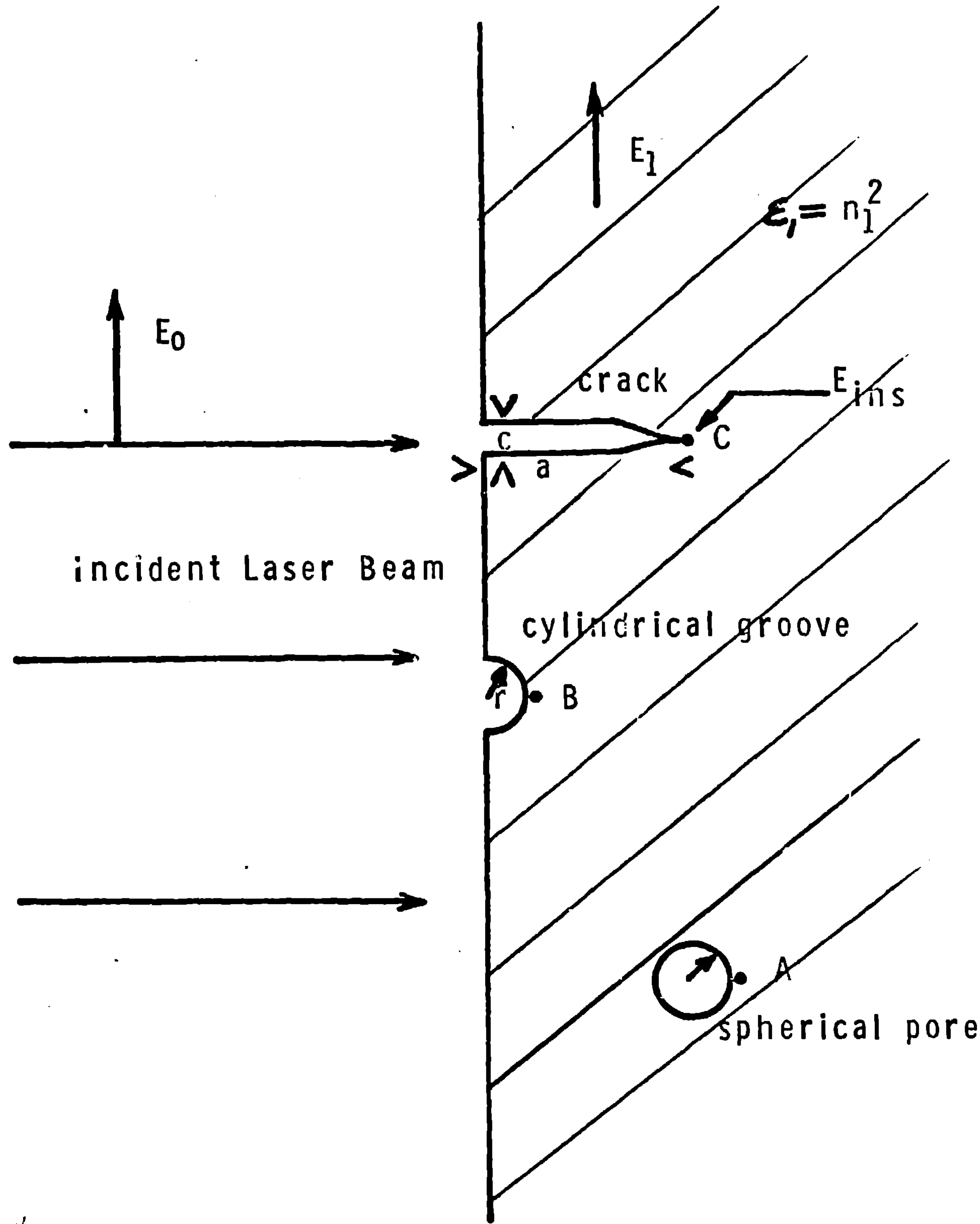
Fig. 5. Damage threshold versus CO_2 laser pulsewidth for AR-coated and uncoated Ge. Spot size = 1.1 mm. (after Newnam and Gill)

Fig. 6. Threshold field versus pulse duration for NaCl (bulk). Measurements were made at 1.06 um with a spot-size radius of 9 um (after Bettis).[12]

Fig. 7. Focusability versus number of CO_2 laser pulses transmitted through an AR-coated ZnSe window. Laser pulselength is 1.2 ns and spot-size radius is 1.1 mm (after Gill and Newnam).[16]

Fig. 8. Focusability versus number of CO_2 laser pulses transmitted through an AR-coated NaCl window. Laser pulselength is 1.2 ns and spot-size radius is 1.1 mm (after (Gill and Newnam).[16]

Fig. 9. Effective lifetime of an AR-coated NaCl window versus CO_2 laser fluence. Laser pulsewidth is 1.2 ns. (after Gill and Newnam).[16]



E_1
$\varepsilon_1 = n_1^2$
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crack
E_{ins}
c
C
a
incident Laser Beam
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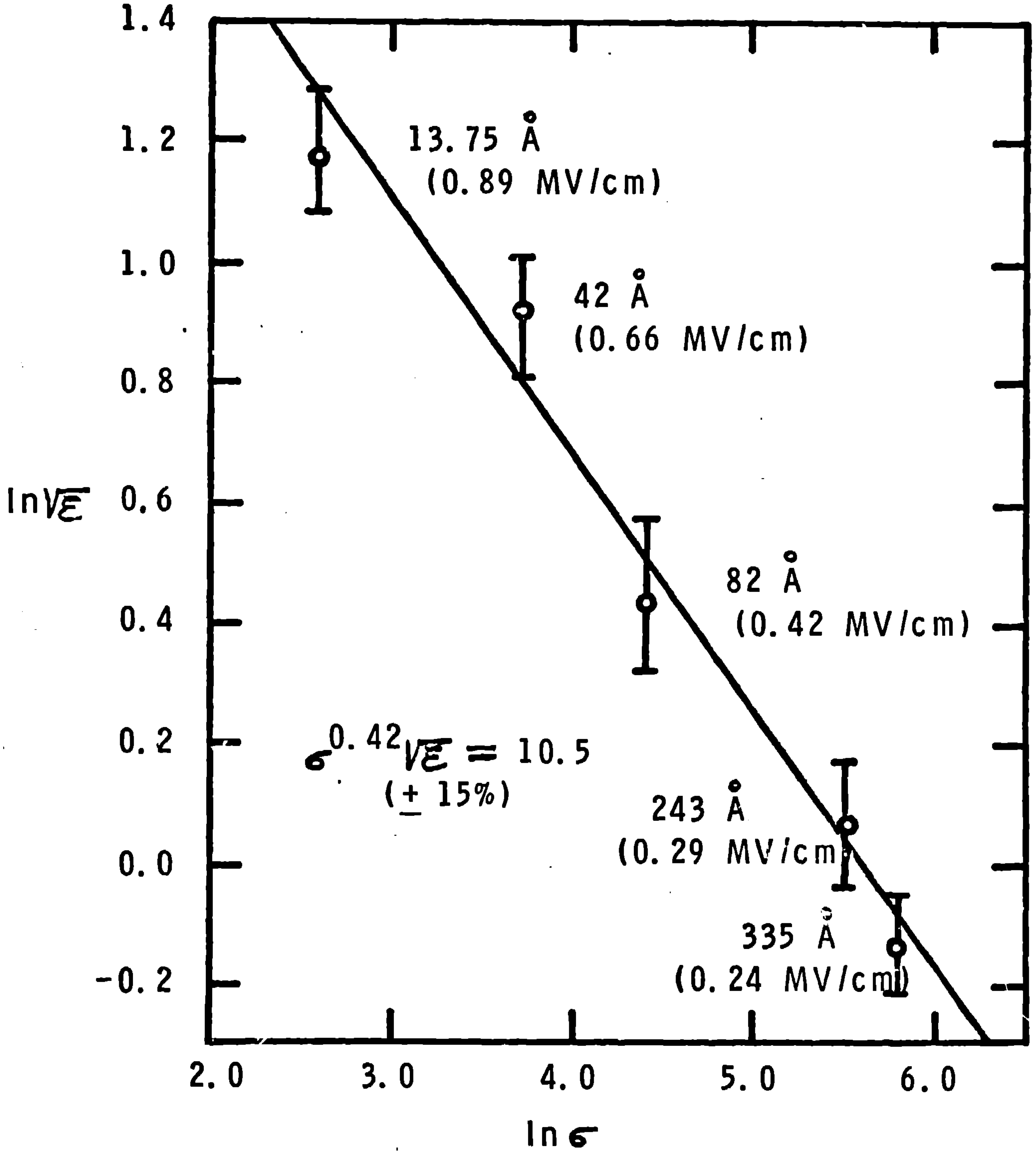
1.4
1.2
1.0
0.8
0.6
0.4
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-0.2
ln√E
13.75 Å
(0.89 MV/cm)
42 Å
(0.66 MV/cm)
82 Å
(0.42 MV/cm)
243 Å
(0.29 MV/cm)
335 Å
(0.24 MV/cm)
σ^0.42 √E = 10.5
(± 15%)
2.0
3.0
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5.0
6.0
ln σ

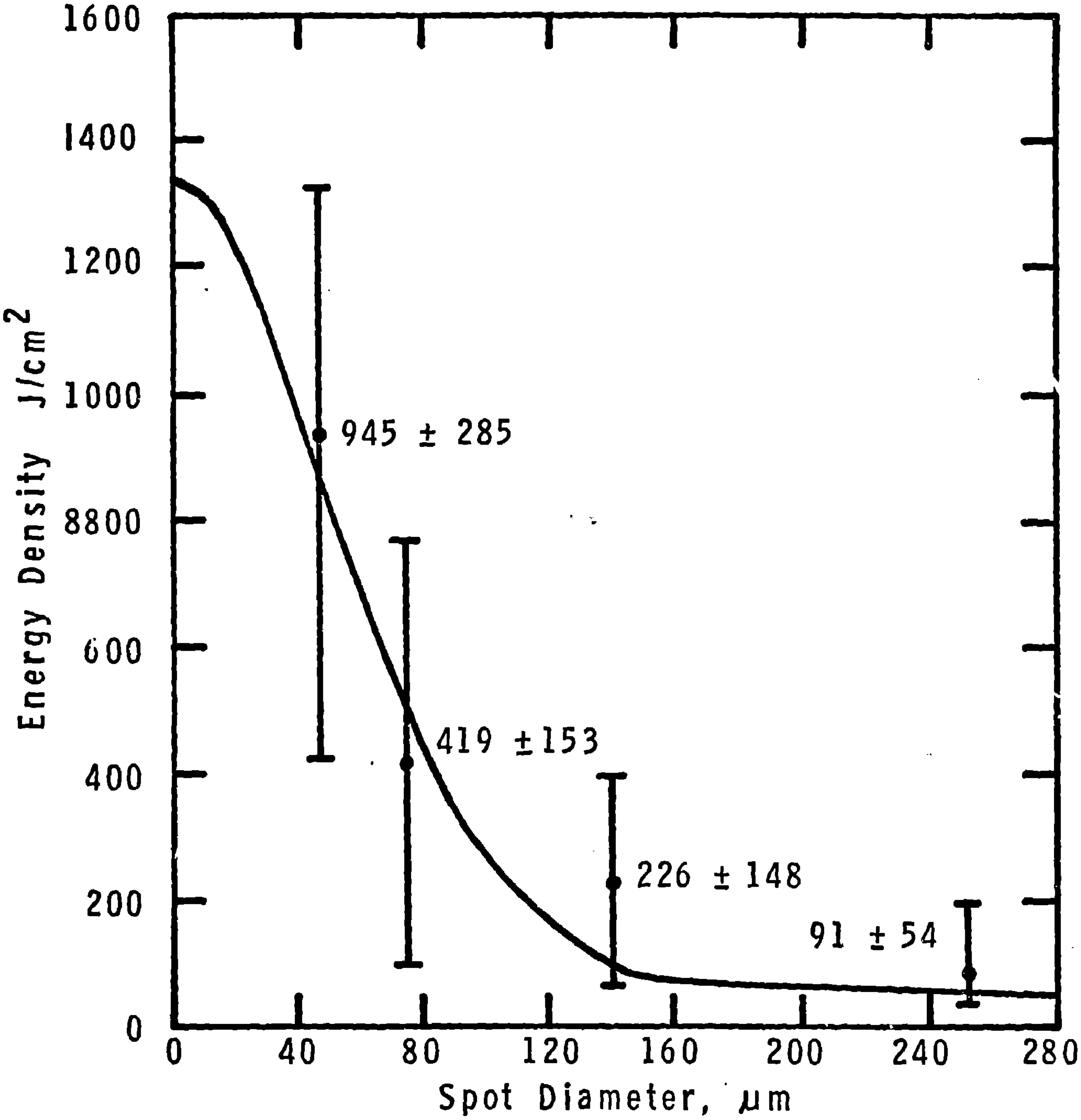

1600
1400
1200
1000
8800
600
400
200
0
Energy Density J/cm^2
945 ± 285
419 ±153
226 ± 148
91 ± 54
0
40
80
120
160
200
240
280
Spot Diameter, µm

Fig 3

STANDING-WAVE FIELDS

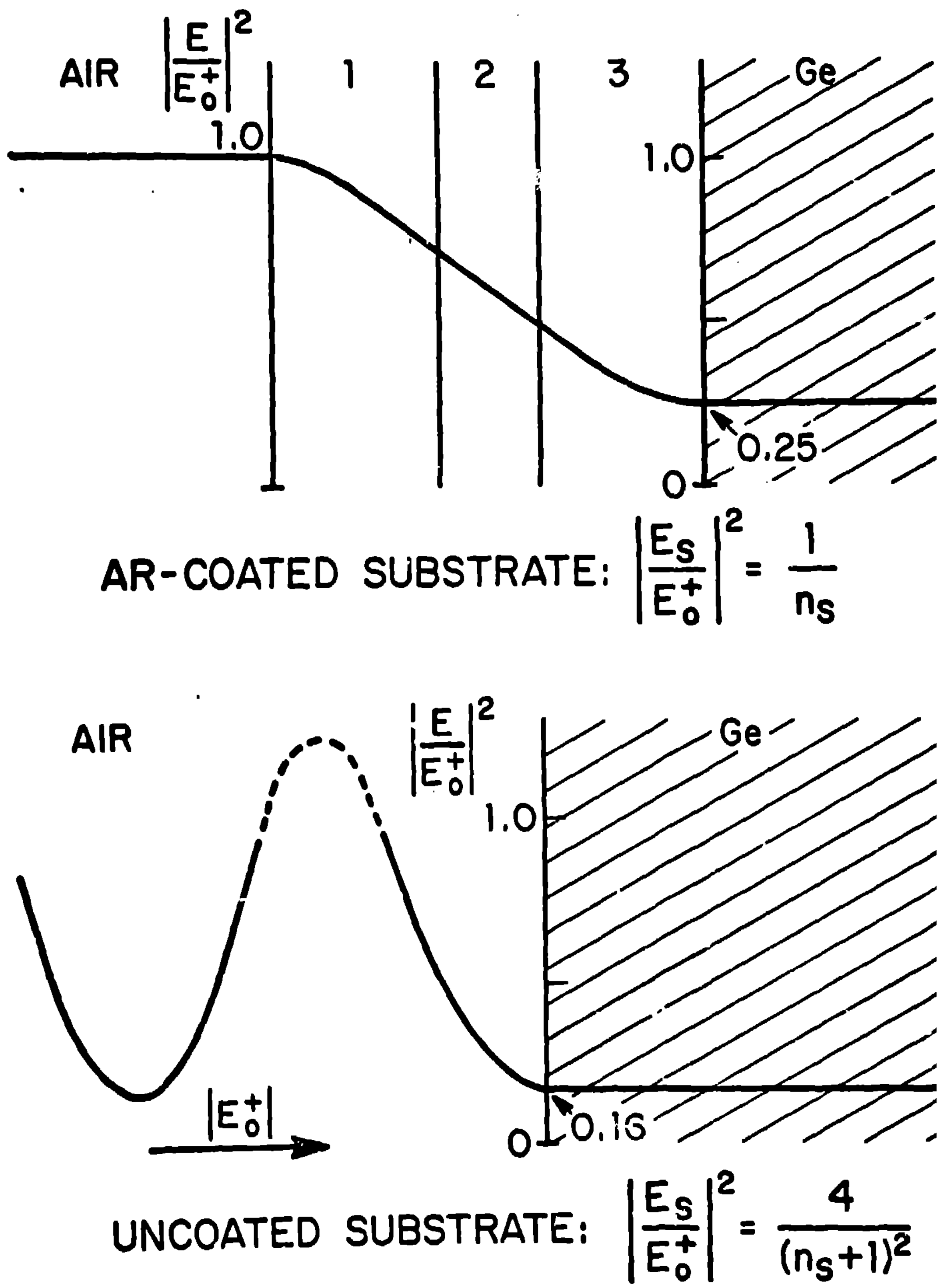
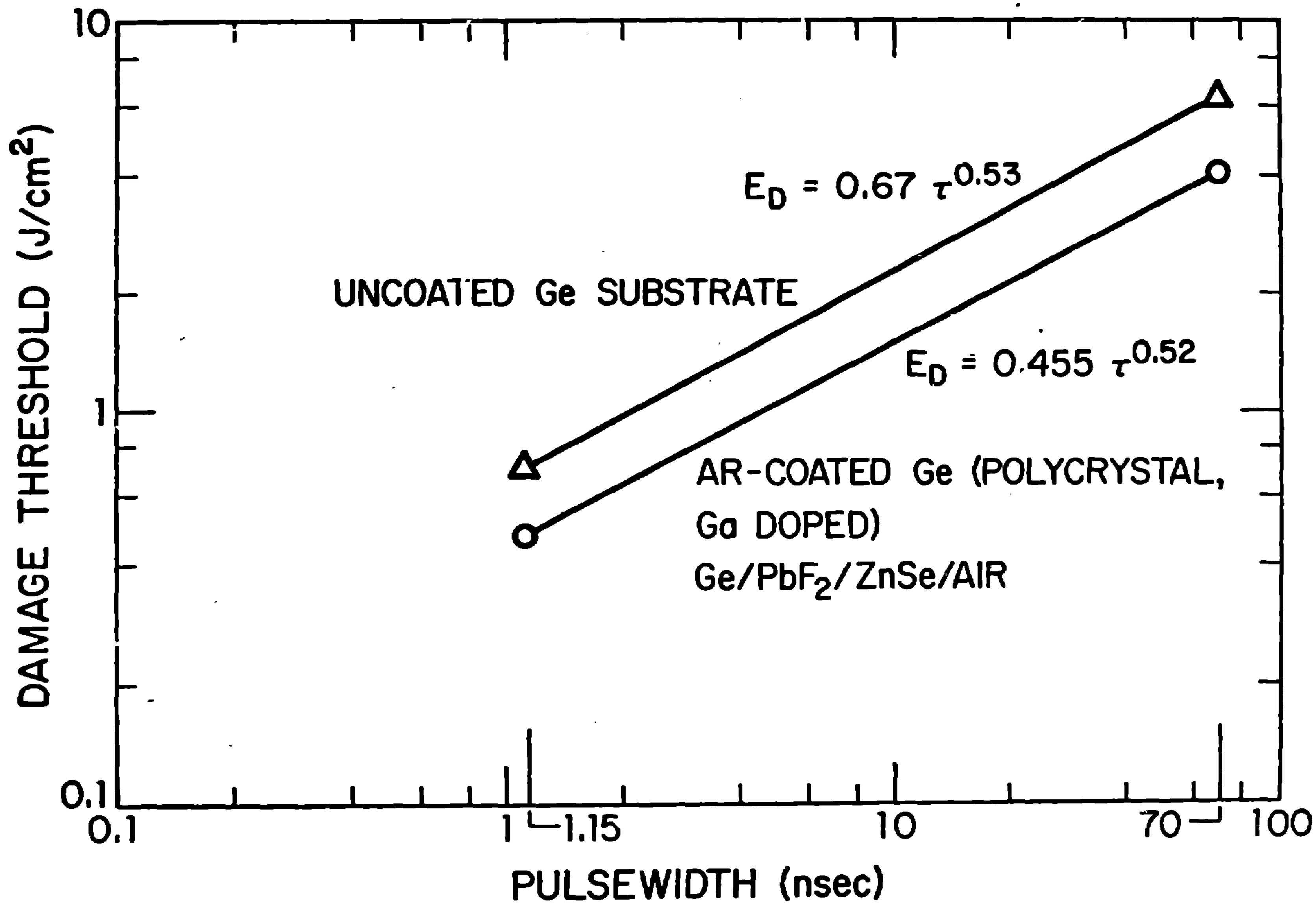
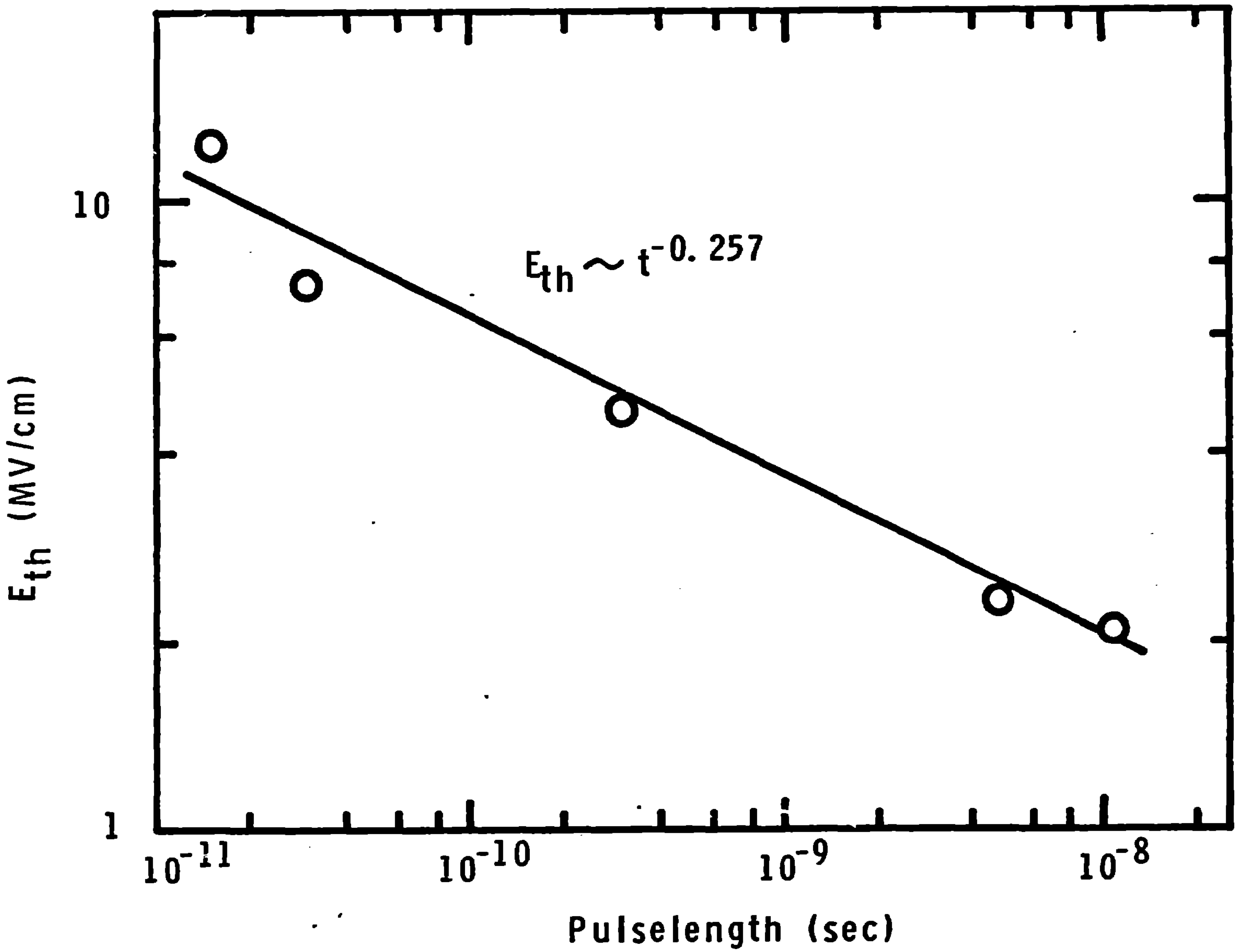


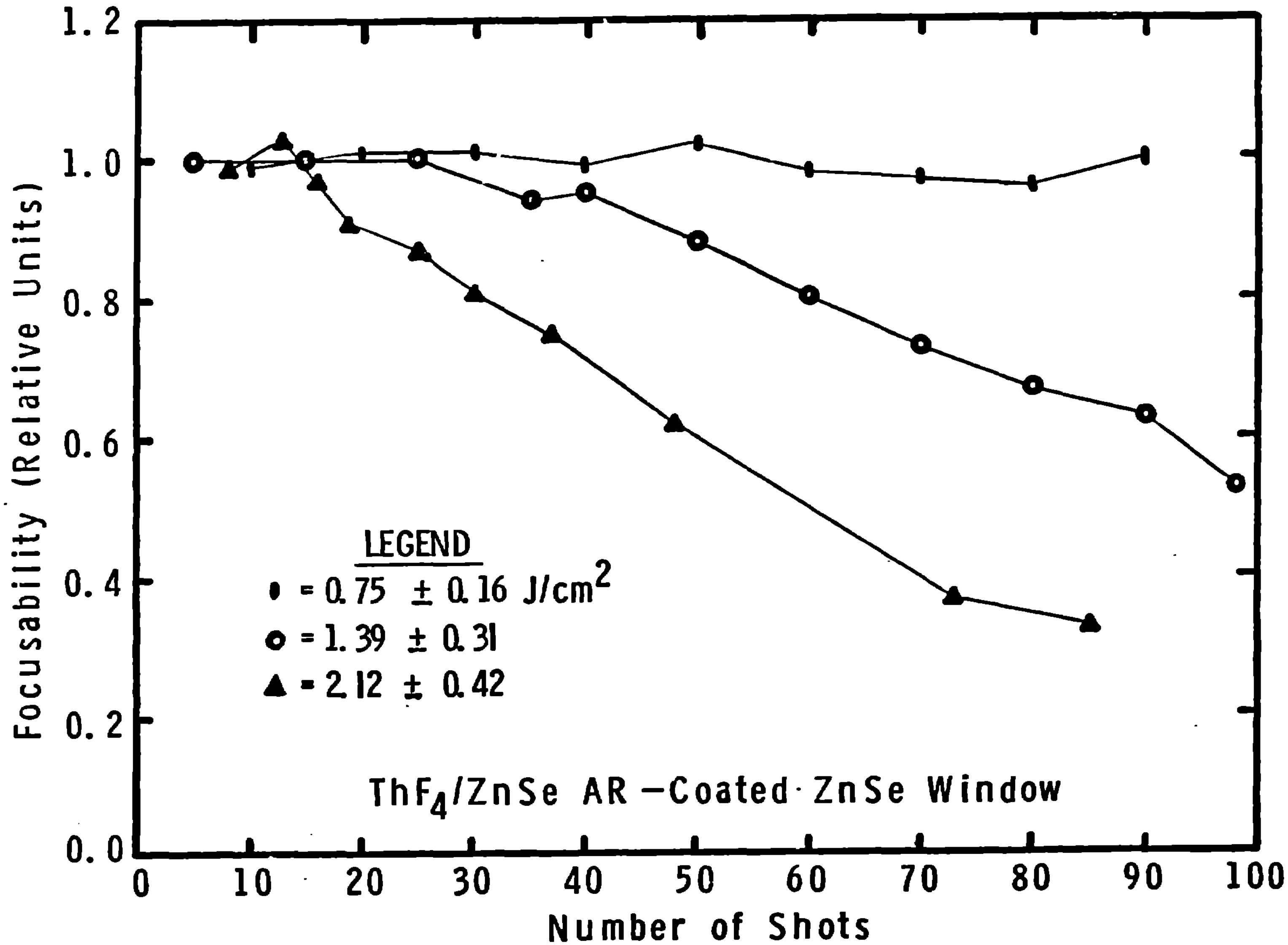
Fig 4



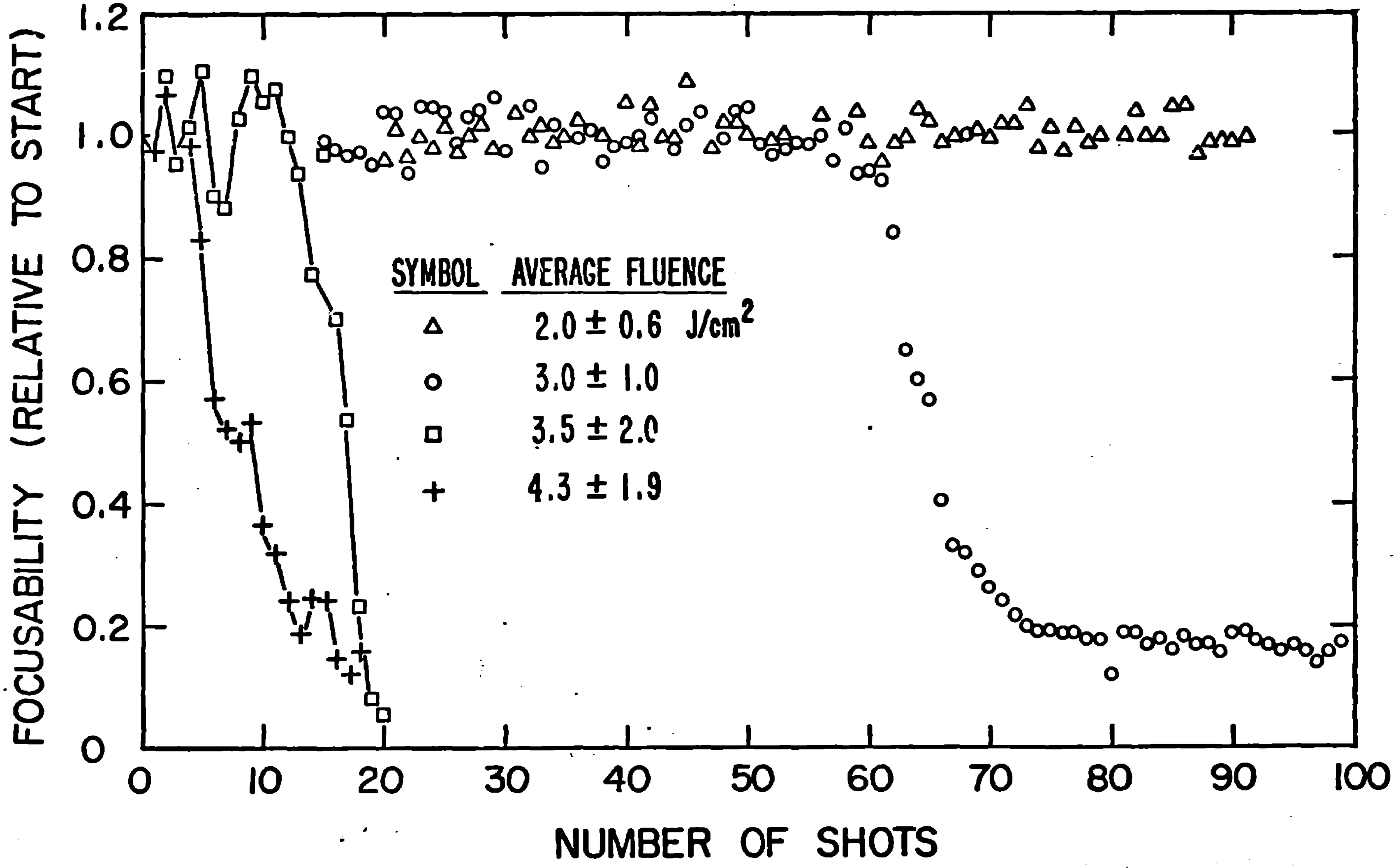
DAMAGE THRESHOLD (J/cm²)
10
1
0.1
0.1
1
1.15
10
70
100
PULSEWIDTH (nsec)
E_D = 0.67 τ^0.53
UNCOATED Ge SUBSTRATE
E_D = 0.455 τ^0.52
AR-COATED Ge (POLYCRYSTAL,
Ga DOPED)
Ge/PbF2/ZnSe/AIR



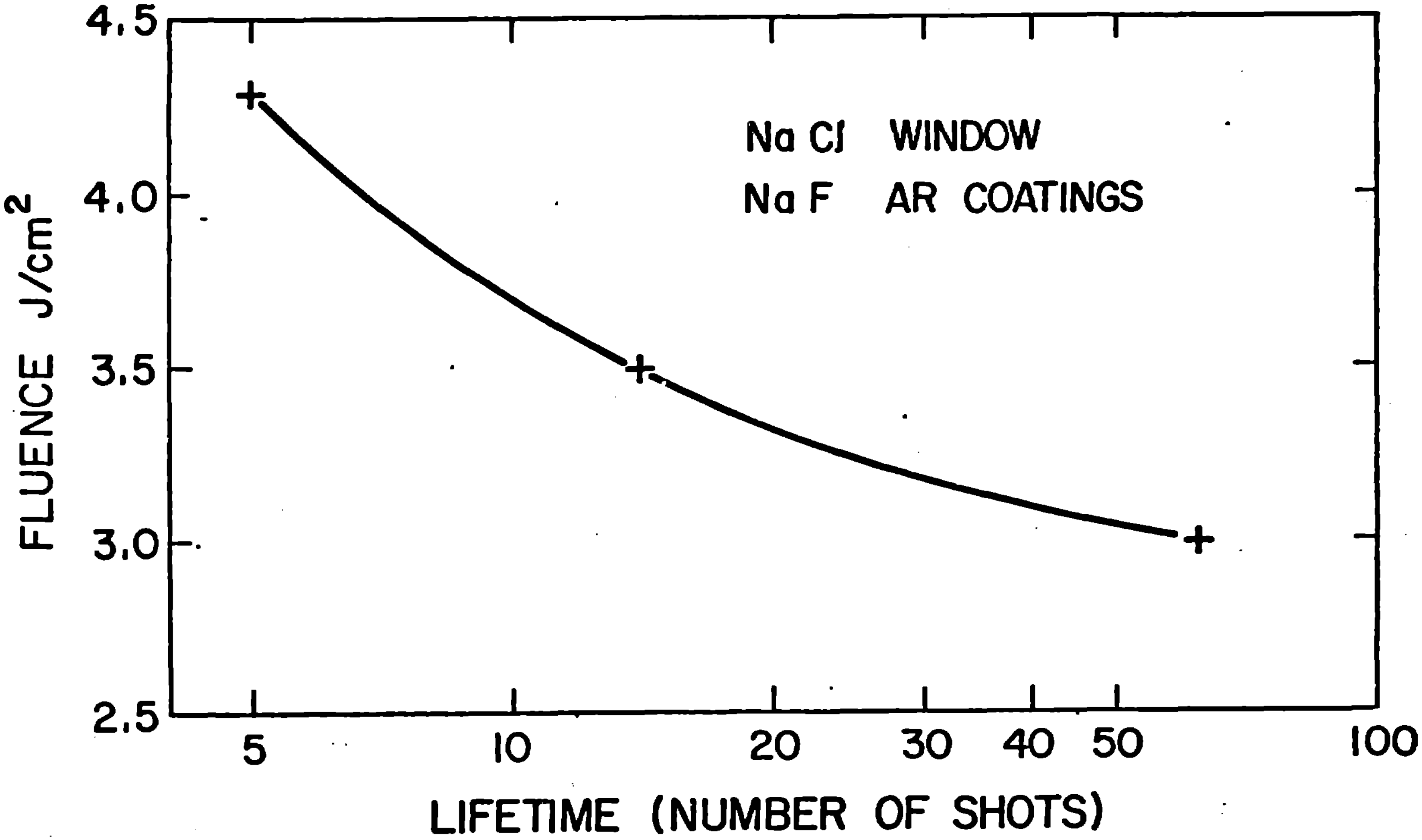
10
1
$E_{th} \sim t^{-0.257}$
E_{th} (MV/cm)
10^{-11}
10^{-10}
10^{-9}
10^{-8}
Pulselength (sec)



1.2
1.0
0.8
0.6
0.4
0.2
0.0
Focusability (Relative Units)
0
10
20
30
40
50
60
70
80
90
100
Number of Shots
LEGEND
= 0.75 ± 0.16 J/cm²
= 1.39 ± 0.31
= 2.12 ± 0.42
ThF4/ZnSe AR – Coated ZnSe Window



FOCUSABILITY (RELATIVE TO START)
NUMBER OF SHOTS
SYMBOL AVERAGE FLUENCE
2.0 ± 0.6 J/cm2
3.0 ± 1.0
3.5 ± 2.0
4.3 ± 1.9
0
0.2
0.4
0.6
0.8
1.0
1.2
0
10
20
30
40
50
60
70
80
90
100



Na Cl WINDOW
Na F AR COATINGS
FLUENCE J/cm²
4.5
4.0
3.5
3.0
2.5
5
10
20
30
40
50
100
LIFETIME (NUMBER OF SHOTS)